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Temperature Dependence of Low Field Switching and Coercive Field in Ferroelectric TGS

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The temperature dependence of the maximum switching current density $j_m=j(t_m)$ has been investigated for ferroelectric triglycine sulfate (TGS) at low fields (1kV/cm$<E_m<$2kV/cm) under almost linearly rising pulses, being $E_m=E(t_m)$ the field value at which the maximum switching current occurs. Increasing temperature above room temperature shows an intermediate zone between the two different switching behaviors already reported, which is intimately connected with the presence of a small bias in the sample. We discuss the meaning of coercive field in a ferroelectric in connection with the observed switching behavior.

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INTRODUCTION

Recent work on electron/ion emission from TGS crystals[1-3] and the related surface discharge plasma induced by spontaneous polarization switching indicates renewed interest in the polarization reversal behavior of uniaxial ferroelectrics.

Most switching experimental investigations in the TGS family crystals have been performed at room temperature. In particular, for high field switching in TGS, Bingelli and Fatuzzo[4] did investigate the temperature dependence of the inverse switching time at a few temperatures in the range from RT to the transition temperature (T≅49°C). For switching at intermediate fields, the temperature dependence of the switching behavior was latter investigated by de la Pascua et al[5]. No detailed investigation of the temperature dependence at the low field switching regime, dominated by sidewise and forward domain wall motion, has been systematically performed as far as we know.

In a previous work[6] we investigated the relationship between the maximum domain wall driven switching current density j_m and E_m (the field value at which this maximum occurs) at room temperature. The field used was an almost linear ramp $E(t) = (E_0/t_0)t$ and we did distinguish clearly two distinct regimes.

1) At very low fields (E_m< 1 kV/cm)

$$j_m = B_1 (E_m - E_{cw1})^{3/2} \qquad (1)$$

where B_1 is a temperature dependent coefficient and E_{cw1} is the threshold coercive field for this regime extrapolated from the experimental data.

2) At low fields (in our case, 1kV/cm< E_m <2 kV/cm)

$$j_m = B_2 (E_m - E_{cw2}) \qquad (2)$$

with another temperature dependent coefficient B_2 and a slightly different value E_{cw2} for the corresponding threshold coercive field, extrapolated for this linear regime.

The coefficients B_1 and B_2 from equation (1) and (2) have the expressions[3]

$$B_1 = \frac{M}{\tau_1}\left(\frac{T_C}{T}\right)\left(\frac{1}{2}\frac{\beta}{\beta_w}\right)^{1/2}\left(\frac{N\mu}{P_S}\right)^{1/2}\left(\frac{1}{E_{S0}}\right)^{3/2} \quad (\mathrm{mA\ cm^{-2}})/(\mathrm{kV\ cm^{-1}})^{3/2} \qquad (3)$$

$$B_2 = \frac{M}{\tau_2}\left(\frac{T_C}{T}\right)\left(\frac{1}{E_{S0}}\right) \quad (\mathrm{mA\ cm^{-2}})/(\mathrm{kV\ cm^{-1}}) \qquad (4)$$

where $1/\tau_1$ and $1/\tau_2$ are the transition probabilities, T_C is the Curie temperature, β and β_w are the mean field coefficients corresponding to the bulk and wall motion switching respectively, N the number of dipoles per unit volume, μ the elementary dipole moment, E_{S0} the saturation field, P_S the spontaneous polarization and M is a time independent but temperature dependent factor

$$M = 4\mu\left(\frac{\pi n N}{A b}\right)^{1/2}\left(\frac{P_S}{N\mu}\right)^{1/2}$$

being n/A the density of prepolarized nuclei at the surface of the crystal and b the the unit cell parameter along the ferroelectric axis. Both coefficients B_1 and B_2 include $1/\tau$, the transition probability for a dipole to switch between the two equilibrium states corresponding to the double potential well.

In the present work we will investigate the effect of temperature on the low field switching behavior of ferroelectric TGS, from 22.5°C to 45°C, relatively near the transition temperature T_C but away from the critical region.

EXPERIMENTAL

The TGS samples, with thickness 0.04 to 0.12 cm, were grown at the Institute of Physics, A. Mickiewicz University, (Poznan, Poland). The crystal chosen for investigation was is a disk 0.12 cm of thickness with a gold evaporated electrode of 5mm in diameter.

An special temperature controlled sample holder was constructed for this aim. The holder was a copper cylinder with three concentric compartments. The inner compartment contained the sample in contact with the electrodes and a thermocouple whose signal was measured by means of a Keithley voltmeter 196 System DMM. Silica gel was placed in this compartment to avoid moisture in the sample. In the intermediate cylinder were located the electric connections from the temperature controller Unipan Thermal type 680. The outside cylinder was used for refrigeration. It must be noted that in spite of the accurate control of this device (about 0.1K) the sample temperature increases slightly during all measure process because the thermal effect of the switching current itself.

A Hewlett-Packard generator, model 33120, was used to get bipolar rectangular pulses amplified with a Kepco model BOP 1000M that transforms the initially rectangular pulses in almost linear rise time signals, at least for the relevant low field region at which the switching current occurs (see ref.6, Fig.1). So, the increment of the field values implies an increment in the signal slope after amplification. Both, the field applied and the crystal response were observed and measured using the Hewlett-Packard oscilloscope model 54603B.

RESULTS AND DISCUSSION

Figure 1 shows t_m^{-1}, the inverse of the time needed for the switching current intensity $j(t)$ to reach its maximum, j_m, as a function of the field value at this time, E_m, for several temperatures. Since the shape of $j(t)$ may be approximated by an isosceles triangle[7] of area

$$\frac{1}{2} j_m t_s \cong j_m t_m = 2P_S$$

it is expected that j_m and t_m^{-1}, that are proportional to each other because the total charge switched is constant, behave in a similar way.

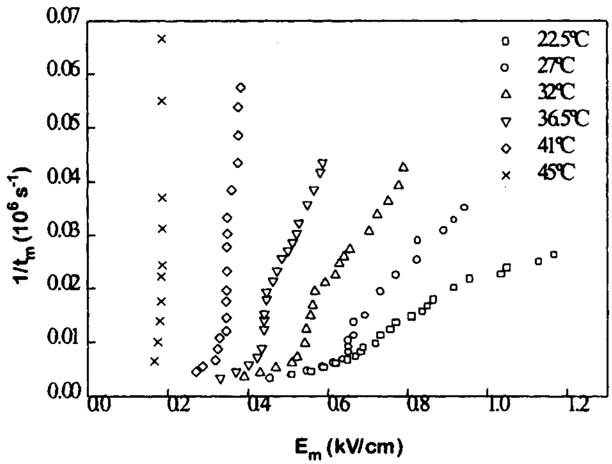

FIGURE 1. Plot of the inverse of the time at which the maximum switching current occurs, $1/t_m$ vs E_m, the corresponding applied field at this moment.

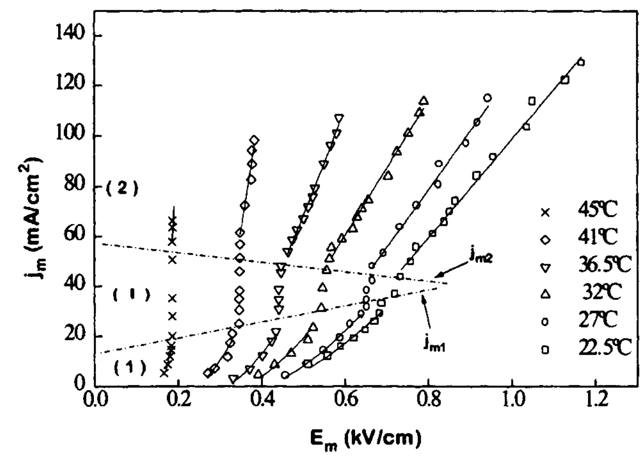

FIGURE 2. Plot of the maximum switching density j_m vs E_m similar to Fig.1. Continuous curves are the fits for the two different regimes: **(1)** with a 3/2 power dependence on $(E_m\text{-}E_{cw1})$ and **(2)** with a linear dependence on $(E_m\text{-}E_{cw2})$. We can distinguish an intermediate zone **(I)** of almost infinite slope between them, which is related to the presence of a bias in the crystal.

Figure 2 plots likewise $j_m(t)$, the maximum switching current attained *vs.* the field value E_m for six different temperatures We can see, as a matter of fact, the proportionality between $t_m^{-1}(E_m)$ and $j_m(E_m)$

For each temperature we can distinguish clearly two different regimes, (1) and (2), with an intermediate zone (I) of almost infinite slope between them that is widening as temperature grows. Their evolution suggests that at RT or lower temperature there would be a clear inflection point between the two regimes[6]. It must be noted that different points in the Figs. 1 and 2 are obtained varying the field pulse height and consequently correspond to different field slopes. So, at the intermediate zone, the different maximum switching current j_m occur at the same value E_m^* independently of the increasing field slope, until reaching the second regime. We call j_{m1} to the minimum value and j_{m2} to the maximum value of j_m corresponding to E_m^*, the field value at the intermediate zone for each temperature. In other words, they are the limiting values of j_m at this intermediate zone.

In the Fig.3 we plot j_{m1} and j_{m2} *vs.* T. The eye guidelines suggest a possible linear behavior and again the tendency to an inflection point at RT. We can check the internal consistency of our description of low field switching by comparing the ratio of j_{m1} and j_{m2} given respectively by Equations (1) and (2) with the experimental values

$$x_{12} \equiv \frac{j_{m1}(E_m^*)}{j_{m2}(E_m^*)} \cong \left(\frac{E_m^* - E_{cw}}{2\beta_w \; P_S^*(T)} \right)^{1/2} \tag{5}$$

where $P_S^*(T)$ is the switched spontaneous polarization at this intermediate zone, which is decreasing as T approaches T_C, and is also related to the relative importance of the bias of the crystal in comparison with the coercive field.

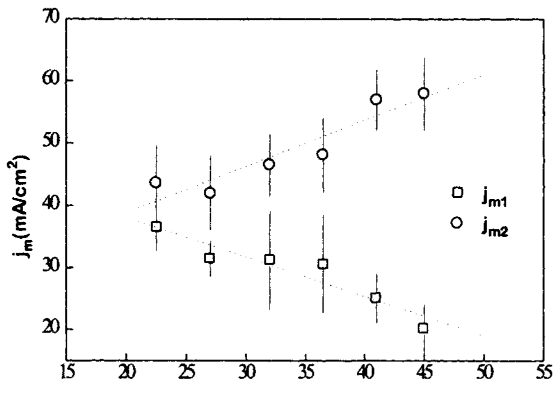

FIGURE 3. Lower (j_{m1}) and higher (j_{m2}) switching current values at E_m^*, the intermediate zone, *vs* temperature T

Table I gives a comparison of $(x_{12})_{cal}$, obtained from Equation (5) with the observed values, $(x_{12})_{obs}$. Taking into account the considerable experimental uncertainties we can see that Equation (5) gives a fair description of the temperature dependence of this intermediate zone at E_m^* (T), which is related to the particular bias in the sample.

TABLE I. Comparison of observed and calculated values of the ratio $x_{12} \equiv j_{m1}(E_m^*)/j_{m2}(E_m^*)$ for various temperatures between 22.5°C and 45°C.

T(°C)	E_m^*(kV/cm)	E_{cw}(kV/cm))	P_S^*(μC/cm^2)	$(x_{12})_{obs}$	$(x_{12})_{cal}$
22.5	0.72	0.44	2.00	0.83	0.88
27	0.66	0.42	1.95	0.74	0.83
32	0.54	0.35	1.60	0.67	0.81
36.5	0.44	0.31	1.25	0.63	0.76
41	0.34	0.25	1.05	0.44	0.69
45	0.18	0.16	0.43	0.34	0.44

Figure 4 displays E_m^* as well as the mean value of the threshold coercive field E_{cw} *vs* T ($E_{cw1} \cong E_{cw2} \cong E_{cw}$). Eye guidelines show that, as it is expected, there is a rapid decrease of both field values as we approach the Curie temperature.

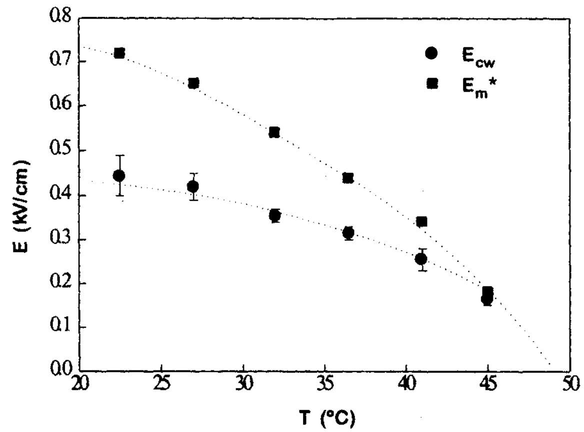

FIGURE 4. Field value E_m^* for maximum switching current, corresponding to the change between regimes (1) and (2), and threshold coercive field E_{cw} given as the mean value of E_{cw1} and E_{cw2} *vs* temperature *T*.

The analysis of switching data as a function of temperature is directly related to the meaning of the coercive field of a ferroelectric crystal. The concept of coercive field is somewhat not clearly defined. It is well known that the direct observation of the half width of hysteresis loops (the conventional definition of coercive field) is strongly dependent on amplitude and frequency or on the slope *dE/dt* as well as on the existence of biasing fields due to impurities or radiation effects. We may note that the switching is incomplete, specially at fields lower than E_m^* as it can be observed on the corresponding hysteresis loops.

We should distinguish

i) The threshold coercive field, E_{cw}, at which appreciable switching begins to appear

ii) The combined field $E_{cw} \pm E_b$ which takes into account the existence of a bias field, E_b in the crystal. This is intimately related to the asymmetry of the ± switching peaks and the corresponding asymmetry of the hysteresis loops

iii) The field corresponding to the maximum switching current, E_m, that would be identical to the classical coercive field if the hysteresis loop is perfectly squared.

iv) The maximum switching current field signaling the change from a lower field switching regime to a comparatively high field switching regime, E_m^*.

In conclusion, increasing temperature results in progressively growing slopes for the two regimes in a like manner as previously observed[4] for high field switching. Between both regimes appears a sharp transition zone that becomes wider with temperature. This must be due to the relative importance of the sample bias field in comparison with the coercive field as temperature approaches T_C.

The results presented on this work partially characterize the temperature dependence $E_{cw}(T)$ and $E_m^*(T)$, but a more detailed investigation would be necessary to ascertain the role of the bias field on the switching process.

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